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Body centered cubic buffer layers for enhanced lateral grain growth of Co/Cu multilayers

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The effect of buffer layers (BLs) on metallurgical microstructure and giant magnetoresistance of Co/Cu multilayers fabricated on them is discussed. The lateral grain size and the magnetoresistance (MR) ratio of multilayers are generally enlarged with changing the chemical composition of BLs toward a limiting concentration, within the range where the solid solution of body-centered-cubic (bcc) structure is formed. A guiding principle for material research for the BLs, which realize flat interfaces with large lateral grain size in the multilayers, is deduced from the correlation between the MR ratio of the multilayers and the surface energy of bcc BLs: the difference between the surface energy of BL (γ_S) and the interfacial energy (γ_{SL}) in Young–Dupré's equation ($\cos\theta=(\gamma_S-\gamma_{SL})/\gamma_L$) should agree with the surface energy of Co layer (γ_L), which is deposited first on the BL. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1543877]

I. INTRODUCTION

In metallic multilayers, showing giant magnetoresistance (GMR) effect, the roughness of heterointerfaces should be minimized to reduce the ferromagnetic ''orange-peel'' coupling between the magnetizations of adjacent magnetic layers[1] and to align them antiparallel at zero field. One of the key issues in obtaining flat interfaces regards controlling the initial growth layer. Lateral grain growth in the flat surface of the initial deposition layer will provide a desirable template for subsequent multilayer growth. One technical candidate for producing this template is the use of buffer layers (BLs) on the substrate. $Fe_{84}Si_{16}$ is reported as an effective BL to facilitate the lateral grain growth of Co/Cu multilayers, and to result in a large magnetoresistance (MR) ratio of them.[2] However, the mechanism of enhancing lateral grain growth of Co/Cu multilayers with the Fe–Si BL has not been fully understood. The only finding is that the Fe–Si BL has body centered cubic (bcc) structure up to 17 at. % Si and no intermetallic compounds in the Fe–Si system play any role on the enhanced lateral grain growth of Co/Cu multilayers.[3] In the present study, we thus investigate the effect of other bcc solid solution BLs on metallurgical microstructure and GMR of Co/Cu multilayers fabricated on them, in order to clarify the mechanism of the enhanced lateral grain growth and to deduce a guiding principle for the material research of effective BLs.

II. EXPERIMENTAL PROCEDURE

Multilayers, of the form sub. /BL 50 Å/(Co 10 Å/Cu d_{Cu})$_{30}$/Cu 20 Å [BL = Cr–Fe, Fe–Ge, Cr–Ni(–Fe)], were deposited on thermally oxidized Si wafers using a magnetron sputtering machine which was capable of pumping gases down to 4×10^{-11} Torr. In order to eliminate the impurity effects,[4–6] the sputtering chamber was evacuated down to less than 1×10^{-10} Torr, and the ultraclean Ar gas, with an impurity level of less than 1 ppb, was then introduced as the process gas. The Cu layer thickness, d_{Cu}, was optimized to make the MR ratio maximum in the so-called ''first peak'' of the GMR oscillation which ranged from 8 to 11 Å. Measurements were performed at room temperature. The chemical composition and crystallographic structure of the BL was determined from x-ray fluorescence analysis and x-ray diffraction (XRD), respectively, by using separately fabricated single layered BL films, having a thickness of 1000–2000 Å. The interfacial roughness, σ, and. lateral grain size, $D_{\text{in-plane}}$, of the multilayers was determined from low-angle XRD profiles[7] and atomic force microscopy images,[2,3] respectively. The MR ratio, measured by a dc four-point probe method, was defined as $(\rho_0-\rho_s)/\rho_s$, where ρ_0 is the maximum resistivity around zero field and ρ_s is the saturation resistivity under the maximum applied field of 13 kOe. $M-H$ loops of the multilayers were measured with a vibrating sample magnetometer. $1-M_r/M_s$, the measure of the volume fraction of antiferromagnetically coupled regions of the Co layers at zero field, where M_r and M_s are the remnant and saturation magnetization, was determined from the $M-H$ loops, subtracting the magnetization of the BLs.

III. RESULTS AND DISCUSSION

Figure 1 shows the $D_{\text{in-plane}}$ and the σ of the multilayers fabricated on the Cr–Fe BLs as a function of the Fe content in BL. The experimentally determined crystallographic phase of the BLs is also shown at the bottom of the figure. When the Fe content is less than 30 at. % or more than 70 at. %, a Cr–Fe BL forms a single phased bcc solid solution, which is indicated as (Cr) or (Fe). The $D_{\text{in-plane}}$ of multilayers markedly increases on the (Cr) -BL with increasing the Fe content of BL toward the phase boundary on σ-CrFe, and it suddenly drops to small value beyond the phase boundary and remains nearly constant on the σ-phase BLs. The $D_{\text{in-plane}}$ then starts to decrease on the (Fe) -BL with increasing the Fe content

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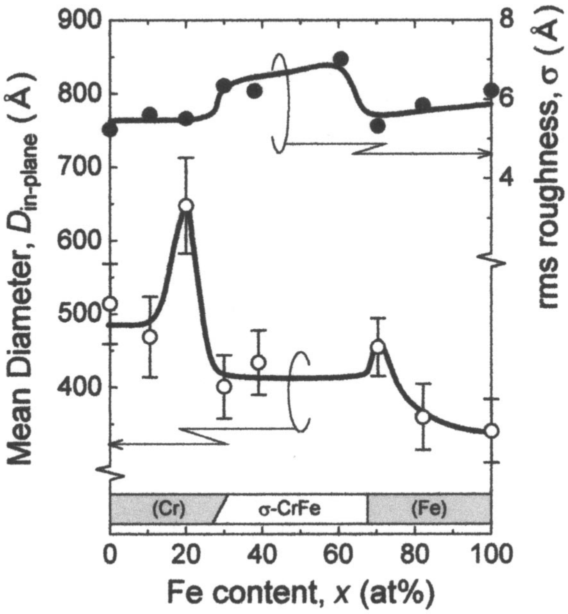

FIG. 1. Changes in mean lateral grain diameter, $D_{\text{in-plane}}$, and root-mean-square roughness, σ, of the multilayers, of the form sub. /Cr–Fe 50 Å/(Co 10 Å/Cu9–10 Å$)_{30}$/Cu 20 Å, as a function of the composition of the Cr–Fe buffer layer.

beyond the phase boundary. On the other hand, the root-mean-square (rms) roughness, σ, of multilayers changes like a step function, corresponding with the change of the crystallographic structure of BLs. The σ does not change significantly in relation to the chemical composition of BLs in the respective BL region: (Cr), σ phase, and (Fe), however, it shows larger value on the σ CrFe BL than on the bcc solid solution BLs. Figure 2 shows the MR ratio and the $1-M_r/M_s$ of the multilayers as a function of the Fe content in Cr–Fe BL. The ρ_s of the multilayers was almost constant for all the Fe contents. The MR ratio and the $1-M_r/M_s$ of multilayers fabricated on the bcc solid solution BLs markedly increase with changing the chemical composition of

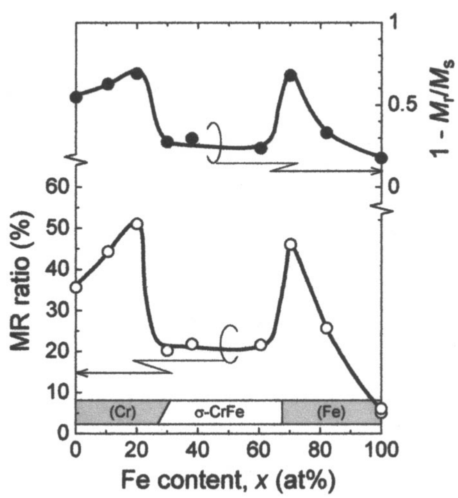

FIG. 2. Changes in MR ratio and $1-M_r/M_s$ of the multilayers, of the form sub. /Cr–Fe 50 Å/(Co 10 Å/Cu9–10 Å$)_{30}$/Cu 20 Å, as a function of the composition of the Cr–Fe buffer layer.

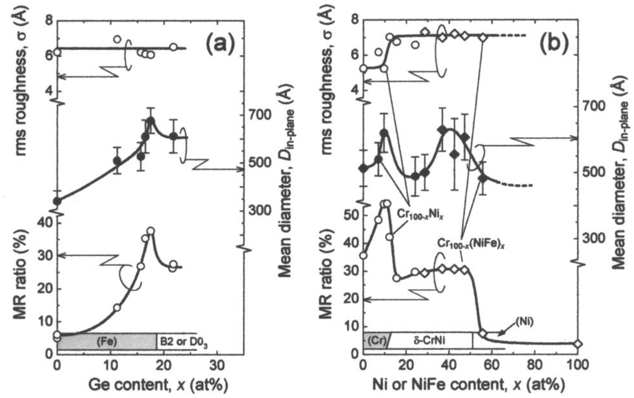

FIG. 3. Changes in MR ratio, mean lateral grain diameter, $D_{\text{in-plane}}$, and rms roughness, σ, of the multilayers, of the form sub. /BL 50 Å/(Co 10 Å/ Cu9–10 Å$)_{30}$/Cu 20 Å, as a function of the composition of the buffer layer (BL). BL=Fe–Ge (a) and Cr–Ni(–Fe) (b).

BLs toward both phase boundaries on σ-CrFe, in coincidence with the changes of $D_{\text{in-plane}}$ and σ. Taking into account that the strength of the orange-peel coupling is given as[1]

$$J_F = \frac{\pi^2 h^2 M_s^2}{\sqrt{2}L} \exp\left(-\frac{2\pi\sqrt{2}d_{Cu}}{L}\right), \tag{1}$$

where h and L are the amplitude and the wavelength of the sinusoidal wave function, characterizing the interfacial roughness, and roughly correspond to the σ and the $D_{\text{in-plane}}$ in the present study, we conclude that the changes in the $1-M_r/M_s$ and resulting MR ratio, shown in Fig. 2, is derived from the change of $D_{\text{in-plane}}$ and σ, through the change of the ferromagnetic coupling strength.

Figure 3 shows the σ, the $D_{\text{in-plane}}$, and the MR ratio of multilayers fabricated on Fe–Ge and Cr–Ni(–Fe) BLs, as a function of the chemical composition of the BLs. The information about the crystallographic phase of BLs is also shown at the bottom of each figure. The MR ratio and the $D_{\text{in-plane}}$ remarkably increase with increasing the Ge or Ni content within the range where the bcc solid solution is formed. The σ shows flat responses against the BL contents within the bcc solid solution regions, and steps up to a larger value when δ CrNi phase is formed. While it is not shown in the figure, the trends of $1-M_r/M_s$ of these multilayers in relation to the BL contents well correspond to those of the MR ratio; consequently, the changes in the MR ratio are similarly derived from the change of $D_{\text{in-plane}}$ and σ in these cases. Focusing on the multilayers on the bcc solid solution BLs in the present study, one finds a general trend in the changes of the $D_{\text{in-plane}}$, while the σ does not change significantly in relation to the chemical composition; namely, the lateral grain size of the multilayers is enlarged and results in a large MR ratio with changing the chemical composition of BLs towards a limiting concentration. This general trend is quite similar to the trend observed in the case of Fe–Si BLs.[2,3]

In order to clarify the physical factors which facilitate the lateral grain growth of the multilayers on bcc solid solution BLs, we plotted the MR ratio of the multilayers against several properties of BLs, such as lattice constant, melting

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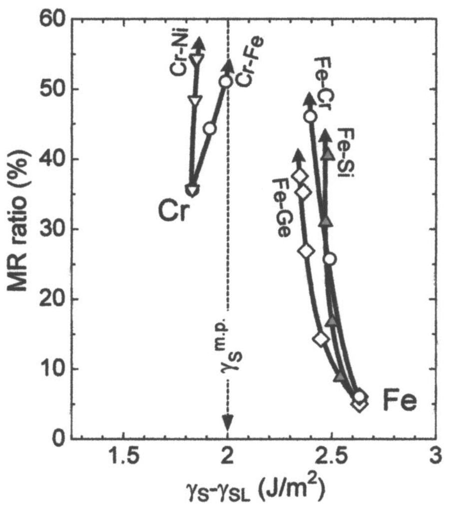

FIG. 4. Correlation between the MR ratio of the multilayer fabricated on body-centered-cubic solid solution buffer layers (BLs) and the surface/interface energy of the BLs.

point, and surface energy. No clear correlation was found between the MR ratio and lattice constant or melting point, but the surface energy of BLs was found to dominate over the MR ratio of multilayers. Figure 4 shows a correlation between the MR ratio of the multilayers and the surface/interface energy of the BLs. The MR ratio only for the multilayers fabricated on the bcc solid solution BLs are re-plotted from Figs. 2 and 3 along the vertical axis. The data for the Fe–Si BL case in Ref. 3 are also plotted. The arrow-heading directions correspond to the increase of the solute contents in the BLs. The horizontal axis corresponds to the energy balance of the surface energy of BL (γ_S) and the interface energy (γ_{SL}) between the BL and Co. The γ_S and γ_{SL} were obtained from Refs. 8 and 9 for pure elements, and were calculated for solid solution alloys by applying a simple diluting distribution function. In Fig. 4, it is clearly found that the MR ratio increases when the $\gamma_S - \gamma_{SL}$ approaches to $2-2.3\ \mathrm{J/m^2}$ from both sides. The surface energy of $2\ \mathrm{J/m^2}$ agrees with the surface energy of solid state Co on melting point ($\gamma_S^{\mathrm{m.p.}}$).[9] Since the MR ratio of the multilayers on bcc BLs is dominated by the $D_{\text{in-plane}}$, as mentioned above, we conclude from Fig. 4 that grains in the Co/Cu multilayers laterally grow large on the BLs which satisfy the condition, $\gamma_S - \gamma_{SL} = \gamma_S^{\mathrm{m.p.}}$.

Assuming that $\gamma_S^{\mathrm{m.p.}}$ corresponds to the surface energy of liquid state Co (γ_L), the peculiar feature in Fig. 4 is discussed by using the well-known Young–Dupré equation about adhesion

$$\cos\theta = \frac{\gamma_S - \gamma_{SL}}{\gamma_L}. \tag{2}$$

The condition, $\gamma_S - \gamma_{SL} = \gamma_S^{\mathrm{m.p.}} (= \gamma_L)$, corresponds to the very limit, at which the angle of contact (θ) becomes zero. Thereby, when $\gamma_S - \gamma_{SL} < 2\ \mathrm{J/m^2}$, the θ of first deposited Co on BL becomes small (in other word, the wettability of Co layer improves) with increasing $\gamma_S - \gamma_{SL}$ and results in large flat islands of Co, which structure is desirable for the large $D_{\text{in-plane}}$ template. This provides a mechanism for the enhancement of lateral grain growth of Co/Cu multilayer on bcc solid solution BLs. However, the decrease of the MR ratio (decreasing $D_{\text{in-plane}}$) with increasing $\gamma_S - \gamma_{SL}$ beyond $2.3\ \mathrm{J/m^2}$ cannot be elucidated within the framework of this mechanism. Namely, according to this mechanism, the enlarged $D_{\text{in-plane}}$ should be obtained independently of the $\gamma_S - \gamma_{SL}$ at the region, because the condition, $\gamma_S - \gamma_{SL} > \gamma_L$, means the spontaneous wetting of Co layer on the BL surface. Further investigation would be needed to answer this discrepancy.

IV. CONCLUSIONS

In conclusion, we deduce a guiding principle for the material research for BLs to realize flat interfaces with large lateral grain size in the multilayers: the difference between the surface energy of BL (γ_S) and the interfacial energy (γ_{SL}) should agree with the surface energy (γ_L) of the layer which is first deposited on the BL.

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